

Strain mediated tri- and quattro- interactions of adatoms.

Wolfgang Kappus

wolfgang.kappus@t-online.de

v0: 2016-10-23

Abstract

Lateral interactions of oxygen adatoms derived from first-principles calculations of the O-Pd(100) system had been claimed to include trio- and quattro terms beside pair interactions. This paper is dedicated to extend an earlier model for substrate strain mediated interactions between adatom pairs to include trio- and quattro terms. While monomers (sitting on high symmetric sites) are supposed to create isotropic stress on the substrate, dimers would create anisotropic stress. The requirement of mechanical equilibrium allows to formulate the elastic energy as a sum of pair-, trio- and quattro terms, solved by respective an eigenvalue problems. Resulting interactions are strongly angle dependent and also reflect the elastic anisotropy of the substrate. A continuum elastic model supports the long range character of the elastic interaction but should also describe some short range features. For the elastically isotropic tungsten substrate a closed solution for quattro interactions is presented, for trio interactions numerical values are given.

1.Introduction

Interactions of adatoms are a subject of continuous interest, various different interaction mechanisms have been described in detail [1]. Strain mediated adatom interactions with the basic s^{-3} distance proportionality and the strong influence of the substrates elastic anisotropy were discussed already in the 1970 decade based on elastic continuum theory [2,3,4]. Also a cutoff method was proposed [3] to handle the s^{-3} singularity, leading to an attractive interaction at very short range.

Elastic interactions appear relevant since many adatom structures reflect the elastic anisotropy of the substrate. Direct measurements of elastic interactions are difficult, so theoretical models seem attractive. Lateral interactions of oxygen adatoms derived from first-principles calculations of the O-Pd(100)

system were claimed to include trio- and quattro terms beside pair interactions [5] which offers an attractive challenge for testing the reach of elastic models. In a parallel study the first-principles calculations are interpreted using long-range strain mediated interactions [6]. The current analysis is dedicated to extend the earlier model for substrate strain mediated interactions between adatoms [3] to include trio- and quattro terms.

In the pair model the interaction of adatoms is mediated by their strain fields generated by single adatoms exerting isotropic stress to their vicinity. Isotropic stress is a consequence of adatom locations on sites with 3- or 4-fold symmetry, selected for simplicity. In a lattice description adatoms would exert forces to their immediate substrate neighbors creating a displacement field equivalent to a strain field.

Basic idea of the current model are adatom pairs stretching or compressing their bond due to their position on substrate sites. Those pairs exert forces to their immediate substrate neighbors and thus create a displacement field to balance the forces. The strain fields of such pairs will mediate interactions between pairs and also between pairs and monomers. The interaction of a monomer with a pair is a trio interaction, the interaction of two pairs is a quattro interaction. Lattice mismatch - known as source of substrate strain [7] - is the mesoscopic consequence of such pair interactions.

If forces between pairs are central, the model can be kept simple and the resulting stress fields can (with symmetry restrictions) be described with one more free parameter than the previous monomer interaction model. The requirement of mechanical equilibrium allows to formulate the elastic energy as a sum of pair-, trio- and quattro terms and to solve them separately.

Calculations below are based on a plain wave expansion of the displacement field and an eigenvalue method, introduced in [8], equivalent to a Green function method used in other papers [7]. Closely following [3] the elastic energy is diagonalized in a set of elastic modes and a generalized eigenvalue problem is established and solved. The elastic interaction will show up as a series of special functions.

The paper is organized as follows:

After outlining the motivation in section 1 the elastic model and its proof are given in section 2. In section 3 the method for calculating interactions is detailed. In section 4 applicability and limitations of the model are discussed. Section 5 closes with a summary of the results.

2. Model details

In this section the overall energy is introduced and the elastic part is diagonalized in terms of eigenmodes. A set of linear equations for the equilibrium displacements in the bulk and at the surface is deduced with an eigenvalue ω to be used later for calculating the elastic interaction of adatoms and dimers. The tensor (and tensor product) notation used below (bold letters) tries to concentrate on the essentials, details of the indices are given in [3]. The text in

the following follows close to [3] but needs extensions to handle the difference between adatom distributions, responsible for pair interactions, and the distributions of adatom pairs, responsible for trio- and quattro interactions. The subscripts k, l will be used to label different distributions. The subscript λ will be used to label eigenmodes.

2.1. Surface energy

The adatom surface energy H is assumed to consist of an elastic part H_{el} and part H_{other} comprising other interactions

$$H = H_{\text{el}} + H_{\text{other}}. \quad (2.1)$$

In the following sections the focus will be put on elastic interactions comprising also short range.

2.2. Elastic energy

Adatoms exert forces on their substrate neighbors leading to a displacement of substrate atoms to balance those forces. Such displacements will increase or decrease the energy of the system. In a continuum description adatoms exert stress parallel to the surface leading to substrate strain which in turn can lead to an attraction or a repulsion of neighboring adatoms. The strength of such strain mediated interaction will depend on the stress adatoms exert on the substrate and on the stiffness of the substrate.

The elastic energy of a substrate with adatoms in a continuous description is given by the sum of two parts, the energy of the distorted substrate and the energy of adatoms exerting tangential forces on the substrate

$$H_{\text{el}} = \frac{1}{2} \int_V \boldsymbol{\epsilon}(\mathbf{r}) \mathbf{c} \boldsymbol{\epsilon}(\mathbf{r}) d\mathbf{r} + \int_S \boldsymbol{\epsilon}(\mathbf{s}) \boldsymbol{\pi}(\mathbf{s}) d\mathbf{s}. \quad (2.2)$$

Here $\boldsymbol{\epsilon} = [\epsilon_{\alpha\beta}]$ denotes the strain tensor, $\mathbf{c} = [c_{\alpha\beta\mu\nu}]$ denotes the elastic constants tensor, and $\boldsymbol{\pi} = [\pi_{\mu\nu}]$ denotes the stress tensor. The integrals comprise the bulk V or the surface S . The strain field $\boldsymbol{\epsilon}(\mathbf{r})$ is related to the displacement field $\mathbf{u}(\mathbf{r})$ by

$$\epsilon_{\alpha\beta}(\mathbf{r}) = \frac{1}{2} (\nabla_\alpha u_\beta(\mathbf{r}) + \nabla_\beta u_\alpha(\mathbf{r})). \quad (2.3)$$

If adatoms do not interact directly with their neighbors, the stress field $\boldsymbol{\pi}(\mathbf{s})$ depends only on the configuration of single adatoms. In case where adatoms do interact with their next neighbors directly, the stress field $\boldsymbol{\pi}(\mathbf{s})$ depends also on the configuration of adatom pairs, because pairs exert forces to the substrate different from monomers. Introducing k as indicator of np different cases, an adatom configuration creates a stress field $\boldsymbol{\pi}(\mathbf{s})$ superimposed of different types $\boldsymbol{\pi}_k(\mathbf{s})$

$$\boldsymbol{\pi}(\mathbf{s}) = \sum_{k=1}^{\text{np}} \boldsymbol{\pi}_k(\mathbf{s}) = \sum_{k=1}^{\text{np}} \mathbf{P}_k \rho_k(\mathbf{s}) , \quad (2.4)$$

where we have introduced np different types of force dipole / stress tensors \mathbf{P}_k and densities $\rho_k(\mathbf{s})$ of adatom monomers or pairs. On (100) surfaces with adatom positions of fourfold coordinated sites $\text{np}=3$; $\mathbf{P}_1=P_1[\delta_{\alpha\beta}]$ stand for an isotropic monomer stress tensor, $\mathbf{P}_2=P_2[\delta_{\alpha 1}\delta_{\beta 1}]$ and $\mathbf{P}_3=P_3[\delta_{\alpha 2}\delta_{\beta 2}]$ for anisotropic dimer stress tensors. $\rho_1(\mathbf{s})$ then stands for the adatom monomer density distribution, $\rho_2(\mathbf{s})$ for the density distribution of x-directed adatom pairs and $\rho_3(\mathbf{s})$ for the density distribution of y-directed adatom pairs. With Eq. (2.4) Eq. (2.2) reads

$$H_{\text{el}} = \frac{1}{2} \int_V \boldsymbol{\epsilon}(\mathbf{r}) \mathbf{C} \boldsymbol{\epsilon}(\mathbf{r}) d\mathbf{r} + \sum_{k=1}^{\text{np}} \int_S \boldsymbol{\epsilon}(\mathbf{s}) \mathbf{P}_k \rho_k(\mathbf{s}) d\mathbf{s} . \quad (2.5)$$

The strain field $\boldsymbol{\epsilon}(\mathbf{r})$ is determined for given densities $\rho_k(\mathbf{s})$ by the requirement of mechanical equilibrium

$$\delta H_{\text{el}} / \delta u_{\alpha}(\mathbf{r}) = 0, \quad (2.6)$$

which leads to freedom of stress in the bulk

$$\nabla \nabla \mathbf{C} \mathbf{u}(\mathbf{r}) = \mathbf{0}, \quad \mathbf{r} \text{ in } V \quad (2.7)$$

and freedom of stress normal $\mathbf{n}(\mathbf{s})$ to the surface

$$\mathbf{n}(\mathbf{s}) \nabla \mathbf{C} \mathbf{u}(\mathbf{s}) = \sum_{k=1}^{\text{np}} \mathbf{P}_k \nabla \rho_k(\mathbf{s}) . \quad \mathbf{s} \text{ on } S. \quad (2.8)$$

With the help of Eqs. (2.7) and (2.8) Eq. (2.5) can be rewritten neglecting boundary effects

$$H_{\text{el}} = -\frac{1}{2} \sum_{k=1}^{\text{np}} \int_S \mathbf{u}(\mathbf{s}) \mathbf{P}_k \nabla \rho_k(\mathbf{s}) d\mathbf{s}. \quad (2.9)$$

2.3. Elastic interaction

An elastic interaction between adatoms can be derived from Eq. (2.9) with an ansatz relating adatom density and displacement field at the surface

$$\mathbf{u}(\mathbf{s}) = \sum_{l=1}^{\text{np}} \mathbf{u}_l(\mathbf{s}) = - \sum_{l=1}^{\text{np}} \int_S \mathbf{A}(\mathbf{s}, \mathbf{s}') \mathbf{P}_l \nabla' \rho_l(\mathbf{s}') d\mathbf{s}', \quad (2.10)$$

which gives inserted in Eq. (2.9)

$$H_{\text{el}} = \frac{1}{2} \sum_{k=1}^{\text{np}} \sum_{l=1}^{\text{np}} \int_S \int_S \mathbf{P}_k \nabla \rho_k(\mathbf{s}) \mathbf{A}(\mathbf{s}, \mathbf{s}') \mathbf{P}_l \nabla' \rho_l(\mathbf{s}') d\mathbf{s} d\mathbf{s}'. \quad (2.11)$$

With

$$V_{kl}(\mathbf{s}, \mathbf{s}') = \mathbf{P}_k \mathbf{P}_l \nabla \nabla' \mathbf{A}(\mathbf{s}, \mathbf{s}') \quad (2.12)$$

this can be written

$$H_{\text{el}} = \frac{1}{2} \sum_{k=1}^{\text{np}} \sum_{l=1}^{\text{np}} \int_S \int_S \rho_k(\mathbf{s}) V_{kl}(\mathbf{s}, \mathbf{s}') \rho_l(\mathbf{s}') d\mathbf{s} d\mathbf{s}', \quad (2.13)$$

with $V_{kl}(\mathbf{s}, \mathbf{s}')$ denoting the elastic interaction between adatoms (or pairs) of type k at \mathbf{s} and adatoms (or pairs) of type l at \mathbf{s}' .

Fig.1 shows different combinations of adatoms or dimers and interactions. 1.a. monomers with stress tensor \mathbf{P}_1 and interaction via V_{11} . 1.b. pairs with stress tensor \mathbf{P}_2 interacting via V_{22} . 1.c. pairs with stress tensors \mathbf{P}_2 and \mathbf{P}_3 interacting via V_{23} . 1.d. monomer with stress tensor \mathbf{P}_1 and pair with stress tensor \mathbf{P}_2 interacting via V_{12} .

2.4. Eigenfunctions

The interactions $V_{kl}(\mathbf{s}, \mathbf{s}')$ are diagonalized by eigenfunctions $X_\lambda(\mathbf{s})$ and eigenvalues $\omega_{\lambda kl}$ given by

$$\int_S V_{kl}(\mathbf{s}, \mathbf{s}') X_\lambda(\mathbf{s}') d\mathbf{s}' = \omega_{\lambda kl} X_\lambda(\mathbf{s}). \quad (2.14)$$

The eigenfunctions form a complete orthonormal basis

$$\sum_\lambda X_\lambda(\mathbf{s}) X_\lambda(\mathbf{s}') = \delta(\mathbf{s} - \mathbf{s}'), \quad \int_S X_\lambda(\mathbf{s}) X_{\lambda'}(\mathbf{s}) d\mathbf{s} = \delta_{\lambda\lambda'}. \quad (2.15)$$

Expanding

$$\rho_k(\mathbf{s}) = \sum_\lambda \rho_{\lambda k} X_\lambda(\mathbf{s}) \quad (2.16)$$

we find from Eqs. (2.13), (2.16), (2.14), (2.15)

$$H_{\text{el}} = \frac{1}{2} \sum_{k=1}^{\text{np}} \sum_{l=1}^{\text{np}} \sum_\lambda \omega_{\lambda kl} \rho_{\lambda k} \rho_{\lambda l}. \quad (2.17)$$

The eigenfunctions X_λ are called density modes. Each mode λ and each density $\rho_k(\mathbf{s})$ causes its own displacement field

$$\mathbf{u}_k(\mathbf{s}) = \sum_{\lambda} \rho_{\lambda k} \mathbf{u}_{\lambda k}(\mathbf{s}). \quad (2.18)$$

From Eqs. (2.14), (2.12), (2.10) we find

$$\mathbf{P}_k \nabla \mathbf{u}_{\lambda l}(\mathbf{s}) = \omega_{\lambda kl} X_{\lambda}(\mathbf{s}). \quad (2.19)$$

Which gives from Eq. (2.8)

$$\mathbf{n}(\mathbf{s}) \nabla \mathbf{c} \mathbf{u}_{\lambda l}(\mathbf{s}) - \omega_{\lambda kl}^{-1} \mathbf{P}_k \mathbf{P}_l \nabla \nabla \mathbf{u}_{\lambda l}(\mathbf{s}) = \mathbf{0}, \quad \mathbf{s} \text{ on } S. \quad (2.20)$$

Eq. (2.20) together with the bulk condition (2.7), for a single mode written

$$\nabla \nabla \mathbf{c} \mathbf{u}_{\lambda l}(\mathbf{r}) = \mathbf{0}, \quad \mathbf{r} \text{ in } V \quad (2.21)$$

constitutes the generalized eigenvalue problem for the displacement field $\mathbf{u}_{\lambda k}(\mathbf{s})$ associated with the density mode $X_{\lambda}(\mathbf{s})$ according to Eq. (2.19). It turns out from Eqs. (2.20) and (2.21) that $\mathbf{u}(\mathbf{s})$ as given in Eq. (2.10) with

$$A(\mathbf{s}, \mathbf{s}') = \sum_{k=1}^{\text{np}} \sum_{l=1}^{\text{np}} \sum_{\lambda} \omega_{\lambda kl}^{-1} \mathbf{u}_{\lambda l}(\mathbf{s}) \mathbf{u}_{\lambda k}(\mathbf{s}') \quad (2.22)$$

is a solution of Eqs. (2.7) and (2.8). The interaction Eq. (2.12) is then with Eq. (2.19)

$$V_{kl}(\mathbf{s}, \mathbf{s}') = \sum_{\lambda} \omega_{\lambda kl} X_{\lambda}(\mathbf{s}) X_{\lambda}(\mathbf{s}'). \quad (2.23)$$

The interaction $V_{kl}(\mathbf{s}, \mathbf{s}')$ in Eq. (2.23) will in the following section be evaluated by associating the density modes with plane waves.

3. Calculation of eigenvalues and interactions

For the calculation of eigenvalues and interactions the label λ will now be replaced by the wave vector $\boldsymbol{\kappa}$ of plane waves for all combinations of tensors \mathbf{P}_k and \mathbf{P}_l . $\omega_{\lambda kl}$ now reads $\omega_{kl}(\boldsymbol{\kappa})$ and the density modes read

$$X_{\lambda}(\mathbf{s}) = X_{\boldsymbol{\kappa}}(\mathbf{s}) = S^{-1/2} \exp(i\boldsymbol{\kappa}\mathbf{s}). \quad (3.1)$$

3.1. Eigenvalue equation

To solve Eqs. (2.20) and (2.21) the displacement fields $\mathbf{u}(\mathbf{r})$ are expanded in plane waves separately for all combinations of tensors \mathbf{P}_k and \mathbf{P}_l

$$\mathbf{u}_{kl}(\boldsymbol{\kappa}, \mathbf{r}) = \sum_{m=1}^3 A_{m,kl}(\boldsymbol{\kappa}) \mathbf{a}_m(\boldsymbol{\kappa}) \exp(\mathbf{g}_m \mathbf{r}), \quad (3.2)$$

where

$$\mathbf{g}_m(\boldsymbol{\kappa}) = i\boldsymbol{\kappa} + \mathbf{n}_{q_m} \quad (3.3)$$

denotes the sum of wave vector $\boldsymbol{\kappa}$ parallel to the surface and a normal component. The \mathbf{a}_m are normalized eigenvectors. The label m denotes 3 displacement modes. For a given $\boldsymbol{\kappa}$ Eq. (2.21) is a homogeneous linear equation for \mathbf{a}_m

$$[\mathbf{c}\mathbf{g}_m\mathbf{g}_m] \mathbf{a}_m(\boldsymbol{\kappa}) = \mathbf{0}, \quad (3.4)$$

where q_m are roots of the characteristic polynomial in q . The boundary conditions Eq. (2.20) become

$$[\mathbf{c}\mathbf{n}\mathbf{g}_m\mathbf{a}_m(\boldsymbol{\kappa}) - \omega_{kl}^{-1}(\boldsymbol{\kappa})\mathbf{P}_k\mathbf{P}_l\mathbf{g}_m\mathbf{g}_m\mathbf{a}_m(\boldsymbol{\kappa})] A_{m,kl}(\boldsymbol{\kappa}) = \mathbf{0}, \quad (3.5)$$

systems of homogeneous linear equations for $A_{m,kl}(\boldsymbol{\kappa})$ with $\omega_{kl}^{-1}(\boldsymbol{\kappa})$ as parameter, \mathbf{g}_m taken from the solution of Eq. (3.4), and k, l standing for the types of strain fields created by the stress tensors $\mathbf{P}_k, \mathbf{P}_l$. Those eigenvalues $\omega_{kl}^{-1}(\boldsymbol{\kappa})$ finally define the interaction $V_{kl}(\mathbf{s}, \mathbf{s}')$ given in Eq. (2.23)

$$V_{kl}(\mathbf{s}, \mathbf{s}') = (2\pi)^{-2} \int \omega_{kl}(\boldsymbol{\kappa}) \exp(i\boldsymbol{\kappa}(\mathbf{s} - \mathbf{s}')) d\boldsymbol{\kappa}. \quad (3.6)$$

3.2. Cosine expansion and integration

To make further use of the results of Eq. (3.6) $\omega_{kl}(\boldsymbol{\kappa})$ is expanded in terms of a cosine series

$$\omega_{kl}(\boldsymbol{\kappa}) = |\boldsymbol{\kappa}| \sum_{p=0}^n \omega_{kl,p} \cos(p\phi) \quad (3.7)$$

where ϕ denotes the direction angle between the x- and the $\boldsymbol{\kappa}$ -direction on (100) surfaces. For symmetry reasons p assumes on (001) surfaces the values 0,4,8 in the case of monomer adatoms sitting on fourfold coordinated substrate sites and the values of 0,2,4 in the case of pairs located on symmetry planes. Higher p values are truncated. The interaction Eq. (3.6) reads with Eq. (3.7)

$$V_{kl}(\mathbf{s}, \mathbf{s}') = (2\pi)^{-2} \sum_{p=0}^n \omega_{kl,p} \int \kappa^2 d\kappa \int \cos(p\phi) \exp(i\kappa s \cos(\chi - \phi)) d\phi, \quad (3.8)$$

where $s = \text{Abs}(\mathbf{s} - \mathbf{s}')$ and χ is the angle between the x- and the $\mathbf{s} - \mathbf{s}'$ direction. Performing the ϕ integral we get

$$V_{kl}(\mathbf{s}, \mathbf{s}') = (2\pi)^{-1} \sum_{p=0}^n \omega_{kl,p} \cos(p\chi) \cos(p\pi/2) \int \kappa^2 J_p(\kappa s) d\kappa, \quad (3.9)$$

where J_p are Bessel functions of the order p . The κ integral diverges. We restrict the κ values to the first Brillouin zone by introducing a smooth cutoff function of the order of the inverse lattice constant

$$V_{kl}(\mathbf{s}, \mathbf{s}') = (2\pi)^{-1} \sum_{p=0}^n \omega_{kl,p} \cos(p\chi) \cos(p\pi/2) \int \kappa^2 \exp(-\alpha^2 \kappa^2) J_p(\kappa s) d\kappa \quad (3.10)$$

with $\alpha \approx 2\pi/s_0$, s_0 is the substrate lattice constant. Performing the κ integral gives for the interaction between adatom monomers ($k, l=1$) or pairs ($k, l=2, 3$) located at the origin and \mathbf{s} , using polar coordinates (s, χ) for their distance $s = |\mathbf{s}|$ and pair direction angle χ with respect to the (100) crystal axis is given by

$$V_{kl}(s, \chi) = \frac{1}{2\pi} \sum_p \omega_{kl,p} \frac{\cos(p\chi) \cos(\frac{p\pi}{2}) \Gamma(\frac{p+3}{2}) s^p {}_1F_1\left(\frac{p+3}{2}; p+1; \frac{-s^2}{4\alpha^2}\right)}{2^{p+1} \Gamma(p+1) \alpha^{p+3}}, \quad (3.11)$$

where ${}_1F_1$ denotes the Hypergeometric Function, $\Gamma(p)$ the Gamma function, and the $\omega_{kl,p}$ denote coefficients of the cosine series Eq. (3.7).

The shape of the interaction elements U_{kl} depends strongly on sign and magnitude of the $\omega_{kl,p}$. $U_{kl}(s, \chi)$ is finite at small distances s and approaches zero with

$$V_{kl}(s, \chi) = (2\pi)^{-1} \sum_p \omega_{kl,p} \cos(p\chi) \cos(p\pi/2) (p+1)(p-1) s^{-3}, \quad s \gg s_0. \quad (3.12)$$

The eigenvalues $\omega_{kl,p}$ as defined by the linear equations (3.4) and (3.5) are proportional to the product of stress parameters $P_k P_l$ and inverse proportional to the elastic constant c_{44}

$$\omega_{kl,p} \sim P_k P_l / c_{44}. \quad (3.13)$$

3.3. Elastic isotropic substrate

For elastic isotropic substrates ($c_{11}-c_{12}-2c_{44}=0$, Voigts notation) some eigenvalues $\omega_{kl,p}$ can be calculated in closed form [9]. They are listed in Tab.1 together with those calculated numerically for Tungsten as substrate. In the case of pair interaction $k=l=1$ the force dipole tensors are isotropic and the eigenvalues for $p>0$ vanish. In the case of quattro interaction $k=l=2$ the force dipole tensor is anisotropic and directed in the x-direction and the 3 eigenvalues differ. In the case of $k=l=3$ the force dipole tensor is directed in the y-direction and $\omega_{kl,2}$ changes sign. For the case of trio interactions $k=1, l=2$ and $k=1, l=3$ no closed solution has been found. The closed solutions for elastic isotropic substrates are useful for testing the code in the more general anisotropic case.

k	l	$\omega_{kl,0}$	$\omega_{kl,2}$	$\omega_{kl,4}$
1	1	$-A$	0	0
1	2	-0.509	-0.509	0.
1	3	-0.509	0.509	0.
2	2	$-B$	$-A$	C
2	3	C	0	$-C$
3	3	$-B$	A	C

Table 1. Coefficients $\omega_{kl,p}$ (in $P_k P_l / c_{44}$ units) for W(100), legend
 $A = c_{11} / (2(c_{11} - c_{44}))$
 $B = (5c_{11} - 2c_{44}) / (8(c_{11} - c_{44}))$
 $C = (c_{11} - 2c_{44}) / (8(c_{11} - c_{44}))$.

The elastic constants used were $c_{11}=523$, $c_{12}=203$, $c_{44}=160$ GPa. From Tab.1 and Eq. (3.12) we conclude the following long range interactions if $P_1 > 0$ and $P_2 > 0$:

- the pair interaction V_{11} is repulsive
- the trio interaction V_{12} is repulsive in x-, weakly repulsive in 45° direction and attractive in y-direction with half strength
- the quattro interaction V_{22} is strongly repulsive in x-, weakly attractive in 45° direction and weakly attractive in y-direction
- the quattro interaction V_{23} is attractive in x- and y-direction and repulsive in 45° direction.

Short range interactions from Eq. (3.11) differ from the simple long range scheme.

3.4. Elastic anisotropic substrate

For elastic anisotropic substrates Eqs. (3.4) and (3.5) have to be solved numerically for all combinations $\{k, l\}$. The $q_m(\boldsymbol{\kappa})$ are derived in Eq. (3.4) from the characteristic polynomial of 6th degree, keeping the 3 solutions with $\text{Re}(q_m) < 0$. The eigenvalues $\omega_{kl}^{-1}(\boldsymbol{\kappa})$ are then derived from Eq. (3.5). For the Pd(100) example we refer to eigenvalues presented in [6].

4. Discussion

The current paper presents an extension of an earlier theory [3]. While previously the adatoms were treated as monomers interacting via substrate strain, the extension allows accounting for dimer effects on adatom configurations.

The model is based on the theory of elasticity in the substrate and on the lateral stress adatoms apply to the surface. Key assumption is the mechanism by which adatoms and dimers interact. Monomer adatoms sitting on fourfold (or threefold) adatom sites expand or contract the substrate by creating isotropic stress. The well-established effect of substrate strain generated by lattice misfit between substrate and overgrown is approximated by the anisotropic stress

adatom pairs exert to the substrate. The latter mechanism should work in cases where adatom pairs are bound electronically and stretched (or compressed) due to their position on substrate sites.

The current model neglects other interactions like dipole-dipole repulsion.

An elastic continuum model for the substrate is rated inadequate for describing short range effects, but describes well long range effects and elastic anisotropies (strong in case of adatom dimers). The cutoff mechanism used in Eq. (3.9) avoids a s^{-3} singularity and therefore extends the models reach towards smaller distances. A different cutoff mechanism will lead to interactions different at small distances. A hard cutoff e.g. will lead to a damped oscillating interaction.

The restriction to high symmetry adatom locations has the advantage of stress parameter P_k degeneracy. On (100) surfaces $P_3=P_2$ due to equivalence of x-directed and y-directed dimers. On (111) surfaces $P_4=P_3=P_2$ for similar symmetry reasons. This reduces the number of free parameters of the model. The dependence of stress parameter values P_k from coverage could be an issue. Its value for isolated dimers could differ from its value in islands since the bond between adatoms depends on their coordination. Different species of adatoms may show different dependencies which could be analyzed with the help of a density-functional-theory calculation.

A further key assumption is an ideal flat surface, i.e. the absence of steps which are known for their significant attractive or repulsive interaction with adatoms.

Unknown is the magnitude of the stress parameters P_k . This is the reason for presenting the interactions V_{kl} in scaled units only. It is, however, assumed that the qualitative statements on the directional dependencies and on the interactions sign hold.

The tungsten example as substrate material was chosen because tungsten is elastically isotropic and therefore allows to test the code for numerical calculations of the eigenvalues ω . The question, if elastic interactions are relevant for interpreting first principles calculations, is left to a forthcoming paper [6].

7. Summary

A model for substrate strain mediated monomer adatom interaction is extended to comprise dimers. While monomers sitting on high symmetry sites exert isotropic stress to the substrate the stress of dimers is strongly anisotropic which causes qualitatively different strain fields and interactions. The interaction of dimers is denoted quattro interaction, the interaction of a dimer with a monomer is denoted trio interaction. The model is evaluated by establishing and numerically solving elastic plane wave eigenvalue equations. For the elastically isotropic tungsten substrate a closed solution for quattro interactions is given, for trio interactions numerical values are presented.

References

- [1] T.L.Einstein, Interactions between Adsorbate Particles, in: Physical Structure of Solid Surfaces (W.N.Unertl ed.), Elsevier, Amsterdam (1996)
- [2] K.H.Lau, W. Kohn, 1977, Surface Sci. 65, 607 (1977)
- [3] W.Kappus, Z.Physik B 29, 239 (1978)
- [4] K.H.Lau, Sol. State Commun. 28, 757 (1978)
- [5] Y.Zhang, V.Blum, K.Reuter, Phys. Rev. B 75, 235406 (2007)
- [6] W.Kappus, to be published
- [7] P.Müller, A.Saul, Surf. Sci. Rep. 54, 157 (2004)
- [8] H.Horner, H.Wagner, Adv. in Phys. 23,587 (1974)
- [9] W.Kappus, Z.Physik B-Condensed Matter 38, 263 (1980)

Acknowledgement

Feedback to this manuscript would be gratefully acknowledged.

Figures

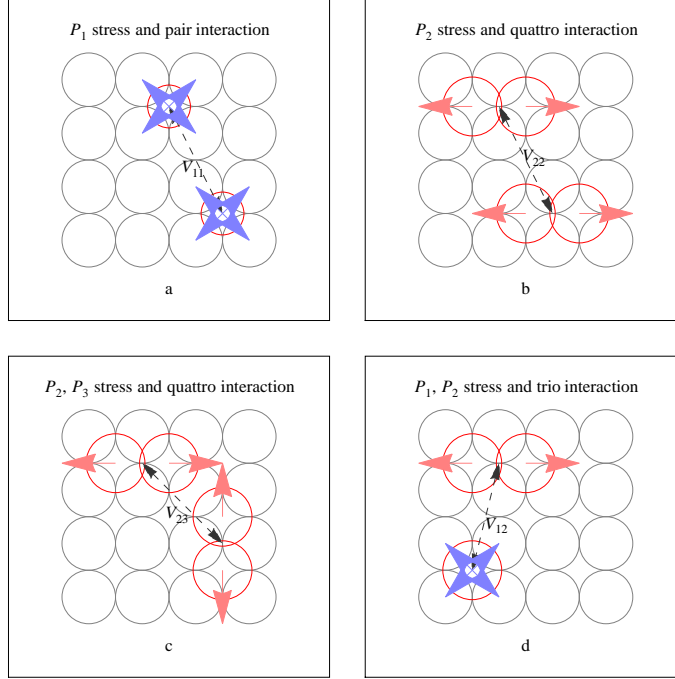


Fig.1: (red) adatoms on (gray) substrate positions with associated stress tensors and interaction types on (100) surfaces. Stress tensors \mathbf{P}_1 are depicted by blue arrows. Stress tensors \mathbf{P}_2 and \mathbf{P}_3 are depicted by red arrows. Black dashed arrows indicate the different interaction types V_{kl} as defined in section 2.

a. monomers with stress tensor \mathbf{P}_1 and interaction via V_{11} . b. pairs with stress tensor \mathbf{P}_2 interacting via V_{22} . c. pairs with stress tensors \mathbf{P}_2 and \mathbf{P}_3 interacting via V_{23} . d. monomer with stress tensor \mathbf{P}_1 and pair with stress tensor \mathbf{P}_2 interacting via V_{12} .

© Wolfgang Kappus (2016)